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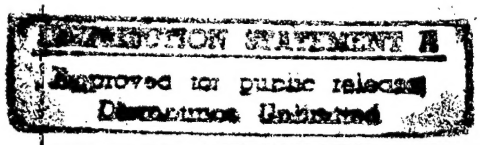

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Abstract

We developed a solid-state tunable mid-IR femtosecond laser system based on a regeneratively amplified Ti:sapphire oscillator. To obtain the desired mid-IR output, a two stage optical parametric amplifier (OPA) was utilized to create two near-IR colors which were subsequently used to produce the desired mid-IR wavelength via difference frequency generation (DFG) in a $LiIO_3$ crystal. Seeding of the OPA was accomplished using the white light continuum produced by the self phase modulation of a Ti:sapphire laser beam focused down into a flow cell containing ethylene glycol. The system design allows for both temporal (i.e., bandwidth) and frequency tuning, affording the user complete output control.

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Final Technical Report for: Femtosecond Infrared Vibration Dynamics Spectrometer for the Study of Supercritical Fluids and Energetic Materials (ONR N00014-95-1-0182)

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TUNABLE MID-INFRARED FEMTOSECOND LASER SYSTEM

Abstract

We developed a solid-state tunable mid-IR femtosecond laser system based on a regeneratively amplified Ti:sapphire oscillator. To obtain the desired mid-IR output, a two stage optical parametric amplifier (OPA) was utilized to create two near-IR colors which were subsequently used to produce the desired mid-IR wavelength via difference frequency generation (DFG) in a $LiIO_3$ crystal. Seeding of the OPA was accomplished using the white light continuum produced by the self phase modulation of a Ti:sapphire laser beam focused down into a flow cell containing ethylene glycol. The system design allows for both temporal (i.e., bandwidth) and frequency tuning, affording the user complete output control.

Ti:Sapphire Oscillator

The first stage of the system is a home-built femtosecond Ti:sapphire oscillator which utilizes a commercial continuous wave (CW) all-lines argon-ion laser as the pump source. The Ar^+ laser is operated at ~4 W in order to obtain a stable pulsetrain in the oscillator. The pumping occurs through a 15 cm focusing lens into a 2 mm Brewster-cut Ti:sapphire rod. Sub-30 fs pulses at 780 nm with a bandwidth of ~45 nm and ~200 mW of power at a repetition rate of 84 MHz are produced via Kerr self mode-locking. The mode-locking is initiated by incurring a small perturbation to the micrometer stage of one of the optics in the oscillator cavity. As self mode-locking is a passive process, no modulation device is necessary to sustain these ultrafast pulses.

Chirped-Pulse Amplification

The system uses chirped-pulse amplification to achieve high peak powers, stretching the pulse prior to amplification and recompressing it post amplification. This is accomplished by sending the oscillator output through a grating stretcher, based on the

design of Martinez, which stretches the pulsewidth to 100 ps. This prevents spectral modification of the pulse during the amplification process and possible damage of optical elements inside the amplifier due to non-linear processes. By using a mask in the stretcher, the spectral bandwidth of the laser beam can be limited to yield temporal pulse tunability (by virtue of the fact that the pulses are transform limited).

Intra-cavity doubled Nd:YAG

Amplification of the stretched seed beam is powered by a 9 W intra-cavity doubled, Q-switched Nd:YAG laser operating at a 1 kHz repetition rate. The intra-cavity doubling is achieved with an 18 mm lithium triborate (LBO) crystal. The design of the cavity is based on a patent by Kuizenga. The problem of thermal lensing is avoided in this cavity design because of a lens system which images the Nd:YAG laser rod onto the doubling crystal. Our cavity layout has been customized to accommodate the damage threshold of the doubling crystal and also utilizes a dual Q-switch design to hold off CW lasing. The output green beam is multi-mode and exhibits a nearly Gaussian spatial profile. The peak-to-peak stability of the output green beam (532 nm) is exceptional, approximately 2-3 %.

Regenerative Amplifier

The doubled beam from the Nd:YAG is focused down to a spot size of 400 μm in a 13 mm Brewster-cut Ti:sapphire rod housed in a home-built regenerative amplifier similar to that reported by Mourou, Barty, et al. The amplifier gain-time and injection synchronization with the Nd:YAG pulse is controlled by custom electronics which operate a Pockels cell purchased from Cleveland Crystals. The seed beam is injected, multi-pass amplified, and then dumped from the cavity ~250 ns (20 round trips) after injection. The output beam and the seed are separated using a Faraday isolator and thin film polarizer combination. The regen output beam is collimated and sent into a grating pair compressor, also based on the design of Martinez. The energy of the resultant compressed beam is >700 μJ at a 1kHz rep rate. By adjusting the slit width of the mask in the stretcher, the pulselength can be varied from 200 fs to 1.5 ps.

White-Light Continuum

Coming out of the compressor, the Ti:sapphire beam is split into three arms of differing energies using a combination of half-wave plates and polarizers. The first arm is used to produce a white light continuum for seeding the OPA. Depending upon the pulse length of the Ti:sapphire laser output (determined by the slit width in the stretcher),

approximately 10-30 μJ of the regen output beam is focused down to a spot size of ~200 μm in an ethylene glycol flow cell using a 10 cm focal length lens. This produces a white light continuum centered about 780 nm (the output wavelength of the Ti:sapphire laser). The stability of the continuum output is highly dependent upon the pump power. As the white light continuum coming out of the ethylene glycol cell is highly dispersive, a 10 cm lens is used to catch the beam and collimate it.

The red wing of this continuum is then used to select the proper near-IR color to seed the OPA. By simply varying the seed wavelength from 1.2 μm to 1.35 μm, it is possible to tune the final output wavelength of the laser system from 2.5 μm to 5.0 μm using $LiIO_3$ as the final difference mixing crystal. By substituting $AgGaSe_2$, it is possible to extend this range to 15 μm. Selection of the seed wavelength is achieved by diffracting the white light off a 600 line/mm grating. Adjusting the angle of the grating thus allows for tuning of the OPA seed color. This in turn allows wavelength tunability of the OPA output colors (signal and idler beams), thereby resulting in a tunable final output color.

Optical Parametric Amplification

The white light continuum generates picojoules of the necessary seed color. Thus, an optical parametric amplification process is necessary to amplify the seed energy to the order of tens of microjoules. As mentioned in the previous section, the bandwidth and color of the seed for the β-barium borate (BBO) OPA is chosen by utilizing a grating to select the correct wavelength out of the white light continuum and diffract it into the BBO crystal. The pump beam for the process is the second arm of the Ti:sapphire compressor output. This 780 nm light is sent down a delay line and through a two lens telescope so that the timing and spot size of the beam can be controlled. The spot size at the BBO crystal is set at ~500 μm so that the power density inside the crystal is sufficient to stimulate optical parametric amplification, but still below the damage threshold of BBO (10 GW/cm^2). A dichroic mirror which reflects the 780 nm pump and transmits the seed (1.35 μm in our current application, which results in the production of 5 μm mid-IR light) is used to combine the two beams and send them into a 7 cm BBO crystal. The portion of the seed which is properly overlapped (both temporally and spatially) and phase-matched with the 780 nm pump beam is then amplified. The BBO crystal currently in use is cut for optimal phase matching (i.e., normal angle of incidence) at 1.35 μm. With a pump beam of 780 nm and a signal of 1.35 μm, an idler of 1.85 μm is produced. The distance of the BBO crystal from the grating in the white light stage, in combination with the bandwidth of the Ti:sapphire output, determines the bandwidth of

the near-infrared output beam. Pumping with 125-150 μJ of 780 nm light, it is possible to obtain ~1 μJ of 1.35 μm light out of the first stage BBO.

The output signal beam of the first stage is then sent into a second amplifier stage in order to further increase the energy. A second 780 nm pump pulse (the third arm of the Ti:sapphire compressor output) is sent along a delay line similar to that of the first pump pulse in order to correctly temporally overlap the signal and pump beams. A 5:1 telescope is used to focus the pump beam into the second BBO crystal and collimate it over the length of the crystal. As in the first stage, the signal and pump beams are combined using a dichroic mirror that reflects 780 nm and transmits 1.35 μm. The spatial overlap of the two beams is adjusted using a combination of mirrors. With a pump beam of >500 μJ and a signal beam of ~1 μJ, the resultant amplified signal beam coming out of the second stage OPA is >20 μJ in energy at 1.35 μm and the idler at 1.85 μm is >12 μJ.

Difference Frequency Mixing

The two beams that are generated in the second stage BBO are frequency mixed in lithium iodate ($LiIO_3$) to attain the desired mid-infrared light (tunable from 2.5 to 5 μm, depending upon the seed wavelength chosen from the white light continuum). However, the group velocities of the signal and idler beams in the BBO are different for the two colors, leading to group velocity walk-off and hence timing mismatch. Another factor which separates the near-IR colors is Poynting vector walk-off, which causes the beams to be askew. In order to achieve efficient optical parametric generation, the beams must be both spatially and temporally overlapped. In addition, for difference frequency generation, the incident beams must have crossed polarizations. However, coming out of the second stage BBO the two colors are of the same polarization. Proper conditions are attained by using a dichroic mirror which reflects one color (1.85 μm) and transmits the other (1.35 μm), thereby separating the two colors. The polarization of the longer wavelength color is rotated with a half wave plate, while timing is controlled by a delay line. The two colors are then recombined using another dichroic beamsplitter and focused into the LiIO3 using a two lens telescope.

With the above design we have achieved energies in excess of 0.7 μJ at 5 μm with a 30 cm^{-1} bandwidth. With recent improvements we hope to obtain energies of greater than 1 μJ, although the output energy is strongly dependent upon the color and pulselength. At shorter pulselengths, the OPA is more efficient due to the high peak power, although this effect is somewhat counteracted due to the fact that the temporal walk-off in the $LiIO_3$ becomes more significant for short pulses. The converse is true at long pulse durations. The shorter the wavelength to which the system is tuned, the

greater the output of the white light stage at the desired seed wavelength, resulting in a higher final output power in the mid-IR. In the future, light of wavelength greater than 5 μm can be produced by replacing the $LiIO_3$ crystal with a $AgGaSe_2$ crystal. This will allow tunability out as far as ~15 μm.

In summary, we have developed an entirely solid-state femtosecond laser system which is capable of producing tunable IR light in the 3-15 μm range. The pulselength of this system can be varied from 200 fs to 1.5 ps (corresponding to a bandwidth range of 10-75 cm^{-1}), allowing us to study both extremely fast processes and vibrational transitions with small anharmonicities. Substantially shorter pulses durations may be produced if desired with little difficulty. With the simple substitution of a $AgGaSe_2$ crystal for the $LiIO_3$ crystal in the final (DFG) stage, it is possible to extend the tunability range of the system out to 15 μm from the current limit of 5 μm. This allows us to access virtually all of the near and mid-IR regions of chemical interest.